

Laser spark ignition: experimental determination of laser-induced breakdown thresholds of combustion gases

Tran X. Phuoc *

Federal Energy Technology Center, P.O. Box 10940; MS: 84-340, Pittsburgh, PA 15236, USA

Received 22 November 1999; accepted 5 January 2000

Abstract

This short paper reports preliminary results on experimental measurements of breakdown threshold laser intensities of air, O₂, N₂, H₂, and CH₄ using a Q-switched Nd–Yag laser operating at 532 nm and 1064 nm and 5.5 ns pulse. The breakdown threshold intensities were measured for the range of pressure from 150 to about 3040 Torr. The results showed that the pressure dependence of breakdown threshold is $I_{\text{thr}} \propto p^{-n}$ which is in agreement with the inverse bremsstrahlung absorption process creating breakdown. The degree by which I_{thr} depends on pressure was found to be stronger at 532 nm than at 1064 nm indicating the important effect of diffusion loss. Published by Elsevier Science B.V.

Keywords: Laser ignition; Breakdown threshold

1. Introduction

In laser spark ignition, when a powerful laser beam of irradiance in the order of 10^{10} W/cm² (or laser photon flux in the order of 10^{29} photons cm⁻² s⁻¹) interacts with a combustion mixture, a spark plasma of high temperature (in the order 10^6 K) and high pressure (in the order of 10^3 atm) is created at the end of the laser pulse. This extreme condition relative to the ambient gas leads to the development of a rapidly expanding shock wave that is of sufficient strength to ignite a gaseous mixture [1–6], liquid fuel sprays [7], or even to extinguish a diffusion flame [6]. It has been shown that the generation

of such a spark is carried out either by the multiphoton ionization process or the electron cascade process [8–12]. In the multiphoton ionization process, a gas molecule or atom simultaneously absorbs a number of photons. If the photon energy absorbed is higher than its ionization potential, the gas molecule is ionized. This process is important only at very short wavelengths (< 1 μm) or at very low pressures (< 10 Torr), where collisional effects are negligible. It becomes insignificant at visible and near-IR wavelengths because the photon energy at these wavelengths is much smaller than the ionization potentials of most gases. For example, the photon energy for a CO₂ laser is 0.1 eV, and for an Nd–Yag laser at 1.064 μm it is 1.0 eV, while the ionization potential for O₂ is 12.071 eV, H₂ is 15.425 eV, CH₄ is 12.51 eV, etc. Thus, the multiphoton ionization process would require the absorption of 120 CO₂ photons (or

* Tel/fax: +1-412-386-6024; e-mail: tran@fetc.doe.gov

12 Nd–Yag photons) to ionize these gases. This is highly difficult.

The electron cascade requires the existence of initial electrons. The electrons then absorb more photons via the inverse bremsstrahlung process. If the electrons gain sufficient energy, they ionize other gas molecules on impact, leading to an electron cascade and breakdown of the gas. At high pressure (≥ 100 Torr) and long wavelength ($\geq 1\ \mu\text{m}$) this process usually dominates the gas breakdown. For ignition application, the creation of a laser spark is usually associated with this process. The initial electrons from which an electron cascade can develop can be generated by the multiphoton ionization process, if the laser irradiance is high enough. The presence of impurities, such as aerosol particles or low ionization-potential organic vapors, can also significantly facilitate the generation of the initial electrons.

Since the laser spark ignition process proceeds first with the formation of a spark, it is of critical importance to determine the laser conditions at which a spark is produced. A knowledge of these conditions is practically important not only for fundamentally understanding the ignition process but also for the selection of lasers, optics windows, and beam delivery system for the design of a practical laser ignition system. Although large amount of studies on the laser-induced breakdown in gases and the rare gases [8–12] has been reported, breakdown thresholds of common combustion gases such as hydrogen, methane, etc., has not been available. In this study, we report some experimental measurements of breakdown thresholds of hydrogen, methane, air, nitrogen and oxygen.

2. Experimental

The experimental apparatus used in this study was described by Phuoc and White [1]. In general, it consists of a single-mode, Q-switched Nd–Yag laser operating at 1064 nm and 532 nm with pulse duration of 5.5 ns. The laser beam was delivered and focused into the ignition cell using a 75 mm-focal length lens. The laser spot diameters were estimated to be $17\ \mu\text{m}$ and $8.5\ \mu\text{m}$ for 1064 nm beam and 532 nm beam, respectively. The laser energy delivered to

the ignition cell was controlled by the laser potential controller and a variable beam splitter that can split the beam from 1% to 99% by rotating about its center. Two pyroelectric energy meters (LaserProbe RJP734) were used for spark energy measurements. One meter was placed behind the exit window facing the incoming laser beam, and the other was placed after the beam splitter which was located before the entrance window. A LaserProbe radiometer (LaserProbe RJ7620) was used to compare the energy levels detected by the two meters. This arrangement allowed the energy meters to detect the transmitted beam through the test cell with and without breakdown.

The threshold of a test gas at a given pressure was measured in the following manner. First, the cell was evacuated and then it was filled with the test gas up to the desired pressure. The laser was fired and its energy transmitted through the test cell was increased until the gas breakdown was observed. The breakdown was easily determined because it was always associated with a cracking noise, the appearance of the bright flash of light in the focal region, and the abrupt absorption of the laser pulse transmitted through the focal region. The threshold breakdown laser energy was measured by the pyroelectric energy meter placed after the beam splitter. The present experiments showed that when high laser energy was used, gas breakdown occurred easily and it was reproducible. When the laser energy was reduced to its breakdown threshold value, gas breakdown became a sporadic event, and the threshold laser energy for initiating gas breakdown could vary by more than 50%. Such sporadic behavior might be due to the difficulty of generating the initial electrons at the breakdown threshold values. The breakdown threshold was then defined as the laser energy at which the gas would break down on more than 50% of the shots.

3. Results and discussions

The present experiments indicate that, for pressure increasing from 150 Torr to about 3000 Torr, laser intensity in the range from 10^{12} to $10^{14}\ \text{W}/\text{cm}^2$ is sufficient to create a breakdown spark in the tested gases. Visible observation indicates that, the sparks

in air, oxygen, and nitrogen were very bright, while those produced in methane and hydrogen had a pinkish color. Fig. 1a to 1e show the breakdown threshold laser intensity for air, O₂, N₂, H₂, and CH₄ as a function of pressure. The breakdown threshold laser energies and laser intensities at 150

Torr and 3040 Torr are also tabulated in Table 1, for easy reference.

The data show that the breakdown threshold laser intensity decreased rapidly as the pressure increased. For pressure increasing from 150 Torr to about 3040 Torr, the breakdown threshold laser intensity for N₂

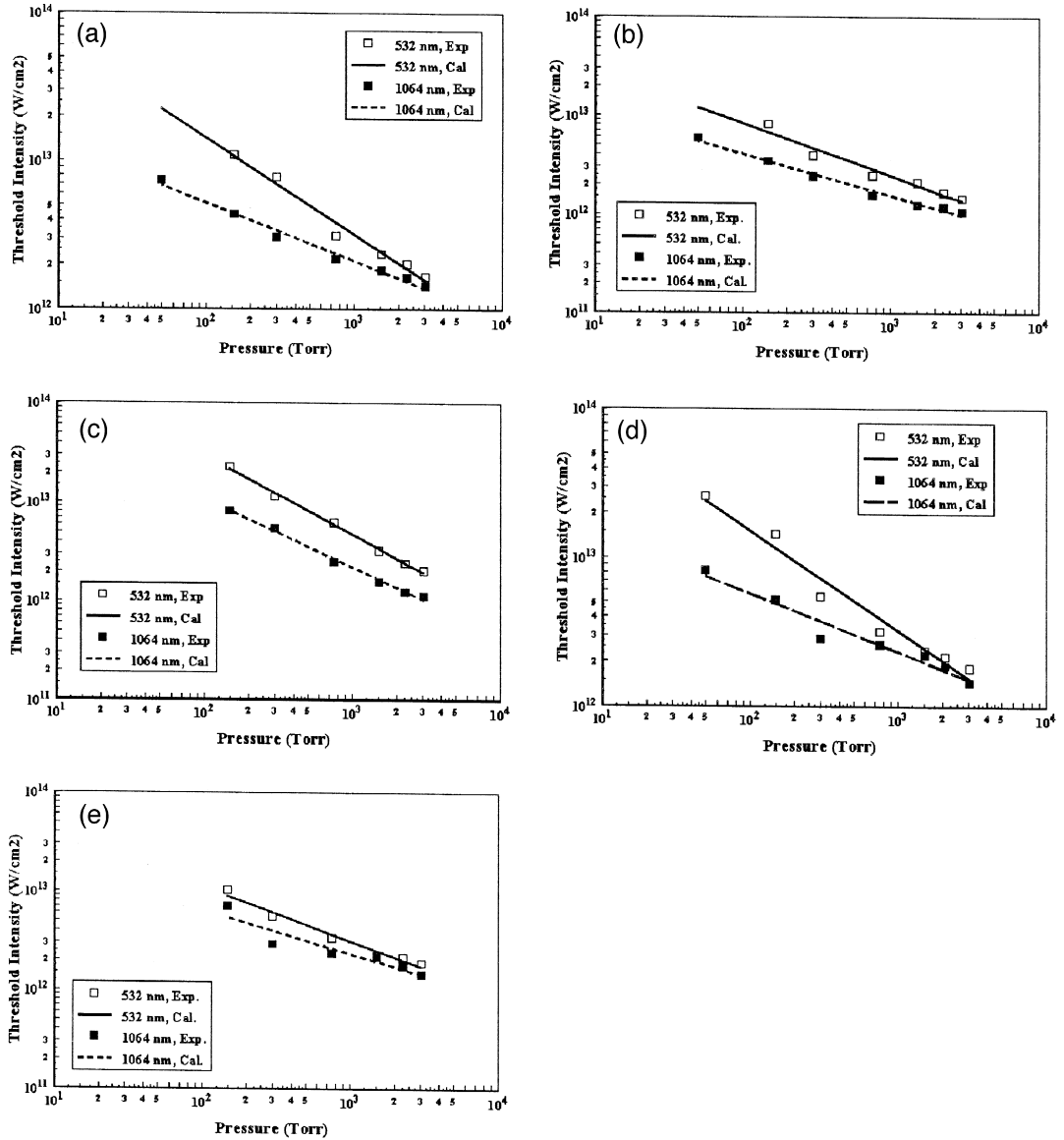


Fig. 1. (a) Breakdown thresholds in air for Nd-Yag laser at 532 and 1064 nm, and 5.5 ns pulse. (b) Breakdown thresholds in CH₄ for Nd-Yag laser at 532 and 1064 nm, and 5.5 ns pulse. (c) Breakdown thresholds in H₂ for Nd-Yag laser at 532 and 1064 nm, and 5.5 ns pulse. (d) Breakdown thresholds in N₂ for Nd-Yag laser at 532 and 1064 nm, and 5.5 ns pulse. (e) Breakdown thresholds in O₂ for Nd-Yag laser at 532 and 1064 nm, and 5.5 ns pulse.

Table 1

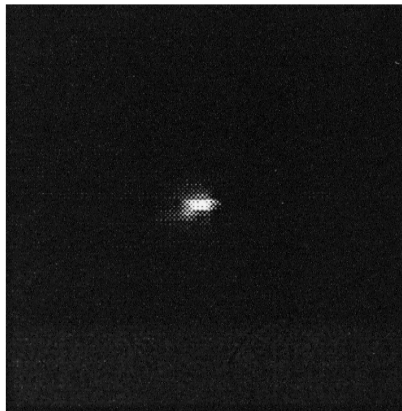
Breakdown threshold laser energy, E_{thr} (mJ), and laser intensity, I_{thr} (W/cm²), in gases (75 mm focal length)

Gas	Pressure	E_{thr} (mJ)		I_{thr} (W/cm ²)	
		532 nm	1064 nm	532 nm	1064 nm
Air	156	20.14	32.31	1.08×10^{13}	4.35×10^{12}
	3040	3.06	10.53	1.65×10^{12}	1.40×10^{12}
CH ₄	150	14.97	25.09	8.05×10^{12}	3.37×10^{12}
	3040	2.67	7.83	1.44×10^{12}	1.05×10^{12}
H ₂	150	42.00	60.00	2.26×10^{13}	8.07×10^{12}
	3040	3.76	8.29	2.03×10^{12}	1.12×10^{12}
N ₂	150	26.37	38.13	1.42×10^{13}	5.13×10^{12}
	3040	3.34	10.74	1.80×10^{12}	1.45×10^{12}
O ₂	150	18.88	51.60	1.02×10^{13}	6.94×10^{12}
	3040	3.45	10.50	1.85×10^{12}	1.41×10^{12}

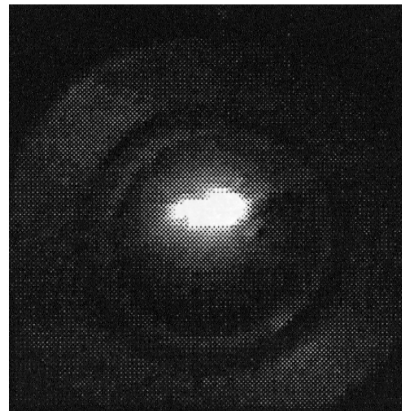
decreased from 1.42×10^{13} to 1.80×10^{12} at $\lambda = 532$ nm, and from 5.13×10^{12} to 1.45×10^{12} at $\lambda = 1064$ nm. For air, it decreased from 1.08×10^{13} to 1.65×10^{12} at $\lambda = 532$ nm and from 4.35×10^{12} to 1.4×10^{12} at $\lambda = 1064$ nm. The breakdown thresholds in H₂ decreased from 2.26×10^{13} to 2.03×10^{12} at $\lambda = 532$ nm, and from 8.07×10^{12} to 1.12×10^{12} at $\lambda = 1064$ nm. These values were found to be in the same order as those of N₂ and about a factor of 2 higher than those of air. The thresholds of methane was found to be consistently lower than those for other gases, it decreased from 8.05×10^{12} to 1.44×10^{12} at $\lambda = 532$ nm and from 3.37×10^{12} to 1.05×10^{12} at $\lambda = 1064$ nm. This is not surprising since methane has an ionization poten-

tial of 12.51 eV which is lower than those of nitrogen (15.58 eV), and hydrogen (15.425 eV). For oxygen, the breakdown threshold laser intensities decreased from 1.02×10^{13} to 1.85×10^{12} at $\lambda = 532$ nm and from 6.94×10^{12} to 1.41×10^{12} at $\lambda = 1064$ nm. Although oxygen has an ionization potential of 12.07 eV, which is the lowest ionization potential compared to those of the gases used, the thresholds in oxygen were found to be in the similar range with those in nitrogen, and hydrogen and about a factor of 2 higher than the thresholds in methane.

Our present data show that the dependence of the threshold on pressure can be expressed as $I_{\text{thr}} \propto p^{-n}$ which is in agreement with the inverse bremsstrahlung absorption process creating breakdown.



Spark in air at 150 torr



Spark in air at 3040 torr

Fig. 2. Typical chapes of laser-induced breakdown in air at 150 Torr and 3040 Torr.

However, the degree by which the thresholds depend on the pressure was found to be strong at shorter wavelength and it became weaker at longer wavelength. At $\lambda = 532$ nm, the pressure dependence of threshold in hydrogen was strongest among the gases used with $n = -0.78$. Breakdown threshold in the other gases showed a weaker pressure dependence with $n = -0.67$ for nitrogen, -0.65 for air, and about -0.55 for both methane and oxygen. At $\lambda = 1064$ nm, the threshold in hydrogen still showed a strong pressure dependence with $n = -0.69$, while the pressure dependence of threshold in other gases became much weaker with n is about -0.4 . Although breakdown of the gases at 532 nm and 1064 nm might be due to the inverse bremsstrahlung process, the observed difference in the degree by which the threshold depend on the pressure might be due to the effect of the diffusion loss out of the focal volume which is more significant at $\lambda = 532$ nm than at $\lambda = 1064$ nm, [11,12].

For a given pressure, our present data show that the breakdown threshold laser intensity at 532 nm was higher than that at 1064 nm. This is in agreement with the wavelength scaling of the breakdown threshold laser intensity discussed by Weyl [13]. In terms of laser energy, however, data in Table 1 show that the breakdown laser energy increased when the wavelength increased. The effect of the wavelength reported here was more profound at low pressure and it became less significant at high pressure. This result indicates that multiphoton ionization process may play a more important role at 532 nm wavelength and low pressure than at 1064 nm wavelength and high pressure. In Fig. 2 we show that shape of spark in air at 150 Torr and 3040 Torr. The shape of the spark in air at 3040 Torr had a cone shape with its large size facing toward the lens. This indicates that the inverse bremsstrahlung absorption process is so dominant that it leads to the development of an absorption front which absorbs the coming photons and propagates toward the laser beam. The spark in air at 50 Torr, however, had a nearly-perfect cylindrical shape and its size is in the order of the focal spot size. There is no sign of such an absorption front propagating toward or away from the lens. This indicates that the multiphoton ionization process is dominant.

4. Conclusions

Laser-induced breakdown thresholds in common combustion gases have been measured using a Q-switched Nd-Yag laser operating at 1064 nm and 532 nm with pulse duration of 5.5 ns. The data show a p^{-n} pressure dependence which is in good agreement with the electron cascade process for creating gas breakdown. For 1064 nm laser beam, except for hydrogen, the pressure dependence was found to be similar for all gas with n is about -0.4 . For 532 nm, the pressure dependence was much stronger showing the important effect of the diffusion loss.

References

- [1] X.T. Phuoc, F. White, Laser-induced spark ignition of CH₄-air mixtures, *Combust. Flame* 119 (1999) 203–216.
- [2] X.T. Phuoc, Multi-Point Laser Ignition of Gaseous Fuels, Multi-Point Laser Ignition of Gaseous Fuels, Proceedings of ETCE/OMAE2000 Joint Conference: Energy for the New Millennium, ETCE2000/ER-10223, New Orleans, LA, February 14–17, 2000.
- [3] T.A. Spiglanin, A. McIlroy, E.W. Fournier, R.B. Cohen, *Combust. Flame* 102 (1995) 310.
- [4] J.X. Ma, D.R. Alexander, D.E. Poulain, *Combust. Flame* 112 (1998) 492.
- [5] E.H. Lim, A. McLlroy, P.D. Ronney, J.A. Syage, in: S.H. Chan (Ed.), *Transport Phenomena in Combustion*, Taylor and Francis, 1996, p. 176.
- [6] R.W. Schmieder, *J. Appl. Phys.* 52 (1981) 3000.
- [7] L.C. Liou, Laser Ignition in Liquid Rocket Engines, 30th AIAA/SAE/ASME/ASEE Joint Propulsion Conference, AIAA-94-2980, Indianapolis, June 1994.
- [8] R. Dewhurst, Comparative data on molecular gas breakdown thresholds in high-laser radiation fields, *J. Phys. D: Appl. Phys.* 11 (1978) 191–195.
- [9] D.I. Rosen, G. Weyl, Laser-induced breakdown in nitrogen and the rare gases at 0.53 and 0.35 μm , *Phys. D: Appl. Phys.* 20 (1987) 1264–1276.
- [10] W. Williams, M. Soileau, E. Van Stryland, Picosecond air breakdown at 0.45 μm , *Applied Physics Letter* 43 (1985) 352–354.
- [11] I.C.E. Turcu, M.C. Gower, P. Huntington, Measurement of KrF laser breakdown threshold in gases, *Optical Communication* 134 (1997) 66–68.
- [12] V.E. Peet, R.V. Tsubin, Multiphoton ionization and optical breakdown of xenon in annular laser beams, *Optics Communication* 134 (1997) 69–74.
- [13] G.M. Weyl, in: L.J. Radziemski, D.A. Cremers (Eds.), *Laser-Induced Plasmas and Applications*, Marcel Dekker, New York, 1989, p. 1.